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Evolving Metallurgical Behaviors in Plutonium from Self-Irradiation

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The plutonium alpha-decay leads to the age-related changes in physical properties. We review our experimental approaches including analytical techniques to assess the effects of extended aging on plutonium alloys, together with our recent results on age-related changes in physical and static mechanical properties. The ultimate goal of this work is to develop capabilities to predict metallurgical evolution driven by aging effects.

Introduction

Plutonium exhibits notoriously complicated metallurgical behaviors, depending sensitively on phase as well as on chemical content and microstructure [1, 2]. Current studies in plutonium metallurgy are motivated by the need to better understand the influence of the metallurgical phenomena on the physical and mechanical properties for stockpile stewardship, nonproliferation, environmental issues, and nuclear power. One of the key areas of research is developing capabilities to predict metallurgical evolution driven by the radioactive decay of plutonium that incessantly creates lattice damage and in-growth of radiogenic helium. Because these integrated aging effects would normally require decades to measure, studies are underway to assess the effects of extended aging on the physical and static mechanical properties of plutonium alloys by incorporating roughly 7.3 atomic % of highly specific activity isotope ^{238}Pu into the ^{239}Pu metal to accelerate the aging process. By monitoring the properties of the ^{238}Pu enriched alloy and naturally aged plutonium alloys, the aging properties of plutonium from the self-irradiation damage can be predicted.

Method

Sample preparation

Radiation damage from alpha decay in plutonium occurs at a rate of ~0.1 dpa (displacement per atom) per year. Because the effects of interest occur over decades, our approach is to accelerate the effects of radiation damage in plutonium metal by incorporating 7.3 atomic % of the higher specific activity isotope ^{238}Pu into the ^{239}Pu lattice. The rate of α-decay of ^{238}Pu is nearly 300 times that of ^{239}Pu so the rate of radiation damage accumulation can be increased. Using this method, the radiation

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damage in plutonium equivalent to sixty years of natural aging can be simulated in only a few years. Additional details of sample preparation are presented elsewhere [3]. In addition, plutonium alloys of various ages are characterized.

Measurement techniques

Details of operation of the dilatometer system, immersion density, and static tensile and compression test techniques are presented elsewhere [3], so only a brief description is provided here. Specifically designed dilatometers, immersion density equipment, static tensile and compression tester were set up inside a nitrogen atmosphere glovebox. The dilatometer is designed to monitor long-term growth resulting from the lattice damage and helium in-growth in plutonium alloys. The immersion density equipment closely matches a design used by Bowman et al. [4] and uses about 200 ml of Fluorinert Electronic Liquid FC-43 as the immersion fluid. The static tensile and compression tester has a specially designed fixture for testing plutonium samples. With a 0.24 inch GL extensiometer for the strain measurement, typical testing was performed at crosshead speed of 0.05 inch/min, so that the ultimate strain rate was about 3.5×10^{-3}/sec.

Results and Discussion

Significant questions remain concerning the metallurgical evolution driven by aging effects. Three principal aging mechanisms have been identified as a result of the self-irradiation of plutonium that would cause metallurgical changes: the initial transient; accumulation of radiogenic helium and actinide daughter products; and void swelling. The initial transient saturates after a short time and results mainly from the increase in lattice parameter. The second contributor to metallurgical changes is the build-up helium and actinide daughters from the radioactive decay of plutonium. The void swelling is another phenomenon, but has not yet been observed in aged plutonium alloys. In addition to these three major mechanisms, a hypothesis suggesting that small precipitates of the higher density ζ-Pu_3Ga phase form in the matrix [5]. Results from dilatometry, immersion density and static tensile measurements show effects from the first two mechanisms on plutonium alloys with ~2 atomic % Ga (see Figures 1 and 2). These techniques are well suited in measuring small property changes produced by aging mechanisms. Results indicate that these plutonium alloys undergo small changes in properties with time, without any signs of void swelling. Aged plutonium alloys exhibit drop in strength when annealed to 300°C indicating the annealing out of the accumulated lattice damage. Current annealing experiments show reduction in the yield strength by ~30 MPa from ~170 MPa on enriched alloys doped with ~3 atomic % Ga (and aged to ~90 equivalent years). This reduction appears to be related to the annealing out the accumulated lattice damage from aged plutonium alloys. We estimate the in-growth of helium contributes ~60 MPa for this alloy aged to ~90 equivalent years.

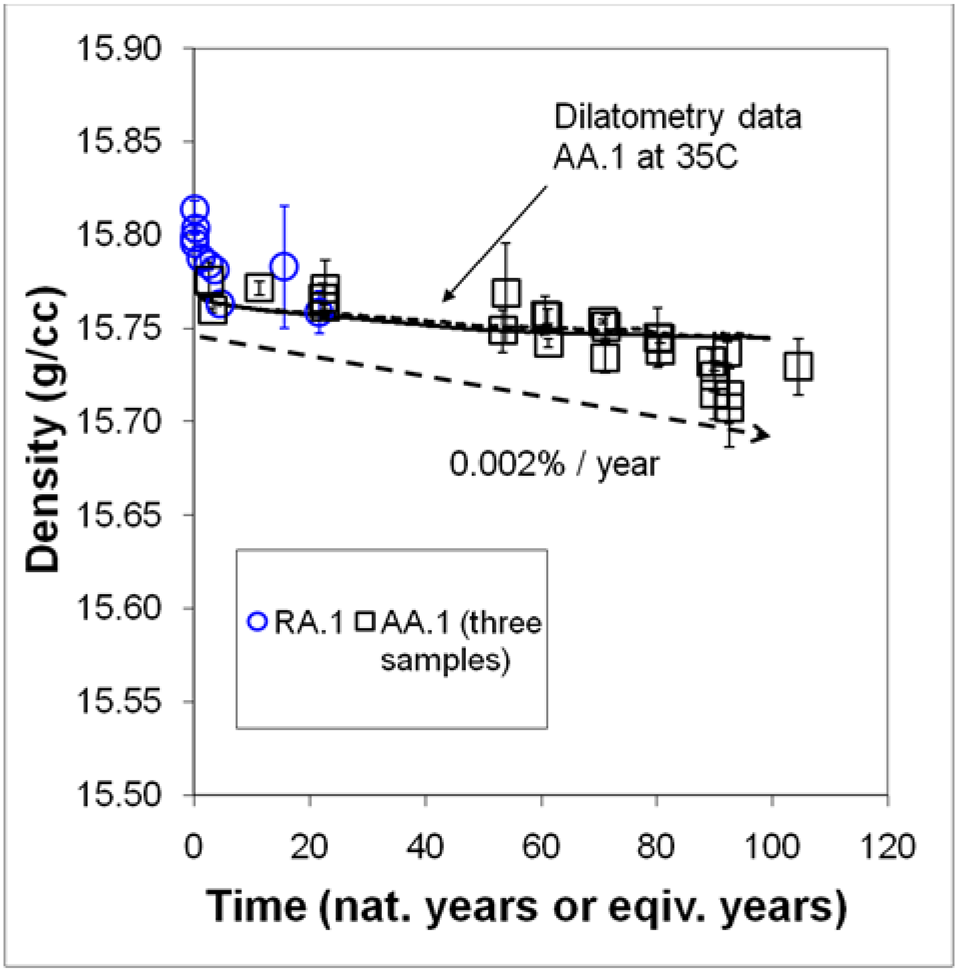

Figure 1. Immersion densities of both enriched (^{238}Pu doped) and reference alloys. The decrease in density is less than 0.002% per year. Circles are reference alloys (RA) and squares are enriched alloys (AA.1). The length change measured by dilatometry is converted to a relative change in density for comparison.

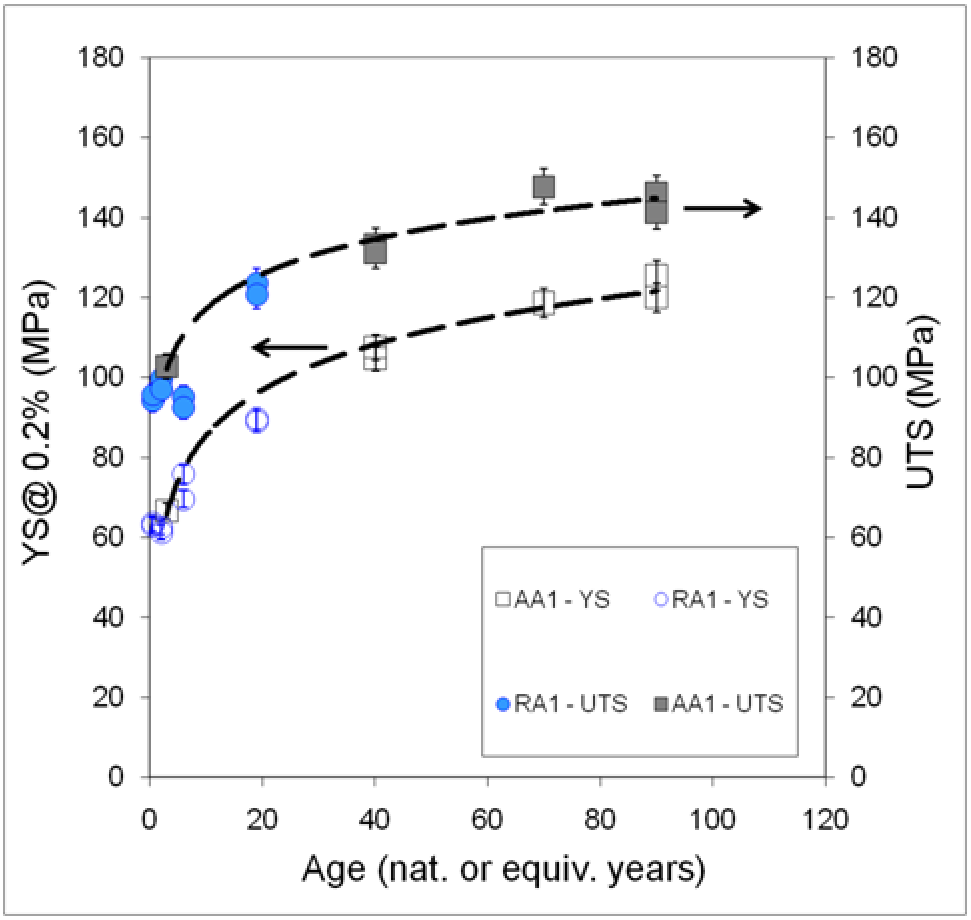

Figure 2. Engineering yield strength (YS) and ultimate tensile strength (UTS) of plutonium alloys from aging [6]. Circles are reference alloys and squares are enriched alloys.

Conclusions

We have developed analytical techniques to measure small changes in plutonium properties by aging. Results of measurements show evolving metallurgical properties of plutonium alloys from incessant self-irradiation damage. So far, however, void swelling has not been observed. Annealing recovery experiments are also under way to better understand aging mechanisms responsible of evolving metallurgical properties with age.

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